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Department of Energy
Nevada Operations Office
P. O. Box 14100
Las Vegas, NV 89114-4100

JUN 10 1987

John E. Rudolph, Director, Program Support, HQ (DP-224) GTN

EXTERNAL REVIEW OF LAWRENCE LIVERMORE NATIONAL LABORATORY (LLNL) DOSIMETRY FOR THE MARSHALL ISLANDS

For quality assurance purposes, I felt it desirable to have an external assessment of LLNL's Marshall Island dosimetry. This review was triggered by the upcoming, independent radiological assessment of Rongelap, the possible return to Bikini and Enjebi by their former residents, and the soon to be convened Claims Tribunal in the Marshall Islands, established under the Compact of Free Association to hear and judge claims arising out of the U.S. nuclear testing program in the Marshalls.

Through the NV Health Physics and Environmental Division (HPE), a Marshall Islands Dosimetry Review Group (MIDRG) was established to assess the current program and make recommendations for future actions, improvements, etc. The comments from the individual members and the recommendations from HPE are attached. Overall, the reviewers found the LLNL dosimetry appropriate and the Brookhaven National Laboratory (BNL) technique for plutonium bioassay worthy of continued support. It was agreed that plutonium is not a major contribution to dose, either by inhalation or from diet. LLNL prospective estimates for Pu have not been contradicted by the retrospective estimates based on the BNL sample measurements.

It was generally agreed a certain amount of monitoring and surveillance would be appropriate in the event populations are reintroduced to Bikini and Enjebi Island. As to the question of the importance and relevance of obtaining autopsy samples of bone and tissue to measure Pu, the conclusion is that it has scientific merit but is a very invasive and culturally undesirable procedure that should not be pursued. The recommendation was also made that the LLNL program at Bikini continue through resettlement to better understand radionuclide transport to the population.

NV's current and proposed actions relative to this review are:

1. Both LLNL and BNL will be sent copies of the MIDRG assessment by each reviewer.

2. BNL has been requested to prepare a five-year plan (fiscal years 1989-1993) radiological safety monitoring program, with costs, for returning populations.

DOE
John Rudolph's Files
D-File
Marshall Islands 1987

3. LLNL has been requested to prepare a five-year plan (fiscal years 1989-1993) environmental sampling and analysis program for Bikini to better quantify radionuclide migration and to refine remedial recommendations.

4. NV will recommend funding the BNL Pu track-etch work at a $150K level in fiscal year 1988 to perform the quality assessment comparisons requested by the MIDRG with no further DP support for that program thereafter. This recognizes that pursuit of this technique is worthy of support, although not in the context of Marshall Islands' programs and resettlement issues. This proposed funding in fiscal year 1988 will allow the work to continue at half the fiscal year 1987 level and allow time for other interested organizations, including other DOE offices, to continue it as they choose.

We will proceed to carry out the above action items pending any DP guidance to the contrary.

/Harry Brown

Harry U. Brown
Assistant to the Manager
for Off-Continent Operations

Enclosures:
As stated

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MAR 3 0 1987

Harry U. Brown, Assistant to the Manager for Off-Continent Operations, NV

HEALTH PHYSICS AND ENVIRONMENTAL DIVISION (HPE) RECOMMENDATIONS ON THE CONTINUATION OF CERTAIN MARSHALL ISLANDS PROGRAMS

At your request, the Marshall Islands Dosimetry Review Group (MIDRG) was established with Kenneth R. Heid, Philip R. Krey, McDonald E. Wrenn, and Bruce B. Boecker as special consultants to review the dosimetry programs of Lawrence Livermore National Laboratory (LLNL) and Brookhaven National Laboratory (BNL) and to make recommendations on the continuance of these programs. The MIDRG met September 30 and October 1, 1986, where William L. Robison and Edward T. Lessard provided information on their respective programs. Reports were received from the four consultants between October 15 and December 15, 1986 (Attachments A, B, C, and D). These comments and recommendations were summarized by David L. Wheeler of my staff in January 1987 and sent to the consultants for review (Attachment E). Two reviewers, Bruce Boecker and Kenneth Heid, have commented on the summary (Attachments F and G). Dr. Boecker's major comment was that the report could be revised slightly into a more coherent presentation. Both reviewers indicated that Mr. Wheeler properly represented the intent of the reviewers in his summary.

The HPE hereby makes the following recommendations concerning the continuation of Marshall Islands dosimetry programs based upon the summary of the comments and recommendations provided by the special consultants.

1. Recommendations regarding the LLNL dose commitment methodology.

There was agreement among the reviewers that the LLNL calculations were appropriate for estimating the environmental transport and uptake of radionuclides in the islands of interest. The LLNL dose estimates for cesium and plutonium should be accepted. The reviewers agreed that the contribution of plutonium toward total dose is insignificant when compared to that contributed by cesium-137.

HPE recommends that LLNL dosimetry calculations be used in regard to resettlement decisions in the Marshall Islands. Following resettlement, there should be a bioassay program that measures both cesium and plutonium to monitor the uptake of radionuclides into the body tissues of returnees. This should continue for a sufficient time to confirm that the actual uptakes are consistent with the LLNL predicted uptakes.

2. Recommendations as to how much further to carry the BNL Pu-in-urine studies.

Dr. Heid recommended that because of the invasive nature of bioassay sampling into the privacy of individuals, the practice of bioassay be continued only until enough data have been collected to establish a baseline by which the environmental transport models can by tested.

Dr. Wrenn and Mr. Krey recognized this technique as a research tool that should be adequately tested against other techniques of comparable sensitivity.

Dr. Boecker agreed with the research utility of the BNL technique of measuring plutonium in urine and suggested that because of the sensitivity of the public toward plutonium in the environment, quantitative measurements of plutonium at environmental levels should seriously be considered by the U.S. Department of Energy.

HPE recommends that the BNL methodology of measuring plutonium in urine by Fission Track Etch be developed and validated against other techniques until it can reliably be used to measure plutonium in urine at environmental levels. This technique should be used for plutonium bioassays recommended in recommendation 1.

3. Is the continuation of the effort worthy of support as a contribution to the state of the art in radiation monitoring and protection?

Drs. Boecker and Wrenn agree that development of the BNL technique has merit for radiation monitoring of the public. Dr. Heid disagrees, stating that it is unlikely that the level of sensitivity offered by the Fission Track Etch procedure will ever be warranted or needed for radiological protection, and Mr. Krey did not express an opinion.

HPE recommends that development of the technique continue for the monitoring of Marshallese following resettlement. The technique is superior to alpha spectroscopy, and it does not make sense for BNL to change to one of the other techniques (still in the developmental stage) for measuring attocurie quantities of plutonium in urine samples. The technique should not be used to extrapolate measured concentrations to hypothesized exposures many years previously.

4. Suggestions for obtaining funding support.

Dr. Heid suggested that the Office of Health and Environmental Research might be willing to assist in support of research involving the collection of autopsy samples. Drs. Wrenn and Boecker suggested the possibility of

assistance from the Department of Defense because of the need for retrospective dose estimates for about 200,000 military personnel. Mr. Krey did not address funding.

HPE recommends that funding continue from Defense Programs until the technique is sufficiently developed and proven to be competitive with other techniques. When this occurs, it will be available for use by the Government of the Marshall Islands and other agencies on a buy-back basis.

5. Can the review group draft a statement as to the importance and relevance to the real Marshall's world of obtaining by autopsy organ and bone tissue representing certain of the unique Marshall Islands population groups?

Three of the consultants were in favor of collecting autopsy tissue and organ samples and measuring the plutonium to evaluate the effectiveness of the bioassay program and the accuracy of the transport and uptake models. Dr. Boecker cautioned against taking autopsy samples unless they are part of a total bioassay program in which organ burdens can be compared with the bioassay results taken from these individuals prior to death.

HPE agrees with Dr. Boecker in that autopsy samples should not be taken unless there are appropriate bioassay data that could be validated by autopsy samples. A long-term study involving bioassay and autopsy samples conducted on specific individuals that have exhibited high levels of plutonium uptake would be valuable. It is not certain that individuals participating in this type of study would benefit personally from the study or that the DOE is willing to make a commitment of this nature. We believe that the information gained from random samples from the population would not have significant merit.

In final response to your concerns about continued funding levels for LLNL and BNL dosimetry projects, we recommend that funding be continued to LLNL until the current programs reach a logical end, resettlement takes place, and the environmental transport of radionuclides to the population is established. Funding should continue to BNL until the fission track etch methodology has been validated by comparison with other techniques and receives credibility by those interested in the measurement of plutonium in urine at environmental levels. The recommendations of the consultants should be provided to the researchers as an aid in accomplishing the desired quality checks on their techniques.

Peter K. Fitzsimmons

Peter K. Fitzsimmons, Director
Health Physics & Environmental Division

HPE/HPB:DLW

Enclosures:
Attachments A-G

October 13, 1986

Mr. Harry Brown
Assistant to Manager for
Pacific Operations
U.S. Department of Energy
P.O. Box 14,100
Las Vegas, NV 89114

Dear Mr. Brown:

I am enclosing a report which contains my evaluation of the data presented at the Las Vegas review of the Marshall Island Program. The report includes both observations and recommendations. As we agreed, I have focused my portion primarily on plutonium dosimetry aspects, although I have briefly discussed certain other aspects that I had opinions on.

Please feel free to call on me if you have any questions or desire further input.

I am also enclosing a billing for my time and for expenses incurred. Thank you for the opportunity to participate in a most interesting program.

Sincerely,

K. R. Heid
Consultant

Enclosure

ACTION A/MOCO
INFO
R.F.
AMA
AME&S
AMO ✓

1. Recommendations Regarding the LLNL Dose Commitment Methodology

The reports presented at the September 30, October 1, 1986 review in Las Vegas indicates that the LLNL staff conducting the environmental survey have established a reasonable program and either have or are getting a good grasp of the environmental conditions in the Marshall Islands and the associated risk to inhabitants. They have characterized the environmental exposure conditions, and the credibility of their data has been established by a good quality assurance program. Based on their data they have concluded that:

- the inhalation mode contributes no more than 1 to 2% of the total dose commitment to occupants at this time,
- the ingestion mode is the primary contributor to the total dose commitment to occupants of the Marshall Islands,
- the contribution to the ingestion dose from transuranic is insignificant when compared to the ingestion dose from radiocesium.

The report prepared by Epidemiology Resources, Inc. based on their 1981-82 review concurred with the conclusions of the LLNL team. Likewise, I find no reason to disagree with the above conclusions. The data do not allow any alternative credible conclusion to be reached. Even during the period of high dust conditions (earth moving tasks, etc.), the air monitoring program did not indicate that inhalation was responsible for any significant intake.

Obviously, it would be nice if better agreement existed between dose estimates based on environmental monitoring and the dose estimates based on bioassay measurements. The two estimates are not as close as one would like at this time, but the difference may be partially explained by the uncertainties involved in predicting their diet. However, even if there were no uncertainties involved, estimates of the dose commitment based on intake via inhalation is always a very questionable procedure, and of the two methods used, the estimates based on bioassay data is much more credible ~~then~~ than estimates based on intake. Despite the differences, there is sufficient correlation between the estimated doses from the two methods at this time to validate the LLNL conclusion that inhalation is

not a major contributor to the committed dose. Further, there is no reason to think that the inhalation dose will increase with time, rather, it should continue to decrease with time as a result of a natural burial in soil process and the radioactive decay of cesium.

The contribution to the dietary dose from plutonium is much less than the dietary dose from cesium (on the order of 10^{-3} to 10^{-4} of the cesium dose) and is dwarfed by comparison with even the uncertainties associated with the cesium dose.

It would seem prudent to continue the environmental monitoring program at the current level for as long as necessary to establish a good data base including a good fix on local diet, contamination levels in locally grown produce, etc. When these baseline conditions have been established, it would seem reasonable to begin reducing the level of the environmental monitoring effort. Even though the ability to estimate dose based on environmental monitoring data is less credible than estimates based on bioassay, the environmental monitoring is less invasive of the Islander's privacy and will monitor exposure condition trends. If an upward trend is observed in the future, the bioassay monitoring program could be accelerated if this seemed necessary.

2. Recommendations as to How Much Further to Carry the BNL Pu in Urine Studies

The bioassay study (in-vivo and excreta) conducted by the Brookhaven National Laboratory is, in my opinion, the only credible way to estimate the dose commitment to the occupants. Thus, the data they have and will collect in the near future undoubtedly will be a major consideration in any resettlement decision. The BNL study has already shown (and the finding has been confirmed by the environmental monitoring program) that plutonium does not contribute significantly to the total dose commitment from either ingestion or inhalation.

However, the level of plutonium activity in the urine samples being collected is very low, and assurance that the results are truly above background is only marginal for most of the samples. In fact, one could even speculate that the small amount of plutonium being excreted in the

urine is the result of recent intake that has been chelated by some preservative in their imported food stuff and as such, did not represent a significant intake. If this were the case, it would not be useful in assessing dose since the model would be inappropriate. However, if assessments were in error because of this, the error would be conservative, that is the dose commitment would be an over-estimation. Another detail that would make the urine excretion more useful, would be to measure the creatinine in the urine to determine the fraction of the total 24 hr urine collected in the sample.

Since the collection of bioassay measurements is more invasive to their privacy, this program should be discontinued as soon as enough data has been obtained to:

- establish baseline of dose commitment being received, and
- verify that a reasonable relationship exists between dose predictions based on environmental monitoring data and dose commitment based on bioassay data.

The possible exception to this might be to continue the collection of urine samples from a select few who have agreed to participate in an autopsy study, if such a study is undertaken. Based on technical considerations only (I can't address the others), it would seem appropriate to select a few subjects who have been excreting at the higher rates and, assuming they are willing to enroll in a registry study to donate partial organ samples at death, continue to collect urine samples for cesium and plutonium analysis as long as positive measurements are being obtained (more on an autopsy study in Number 5).

Discontinuation in a few years of the bioassay monitoring program should be considered only if an adequate environmental monitoring program is in place.

3. Is Continuation of the (Bioassay Monitoring) Effort Worthy of Support as a Contribution to the State-of-the-Art in Radiation Protection?

It is difficult to evaluate the potential impact of the BNL procedure on the state-of-the-art. It is unlikely that the level of sensitiviy

offered by the fission track analytical method being developed by BNL will ever be warranted or needed for routine bioassay monitoring of occupational workers. It likewise is difficult to conceive of any large scale collection of urine samples from members of the public at large. If it were decided that the level of sensitivity provided by their method (~60 aCi/sample) was needed, the radioanalytical procedure would have to compete with other procedures currently in the development stage that might be as sensitive, as cheap to process and may offer faster turnaround time, such as dual laser techniques.

a. Is it Important to Continue to Develop the Fission Track Method of Analysis for Plutonium in Urine in the Marshall Island Context?

The existing data that have been collected to date have all been generated using the fission track analytical technique. Thus, it would seem prudent to continue to analyze samples collected in the future using the same technique. However, I got the impression from data presented at the review that the technique was essentially developed. Some refinement such as a better understanding of the minimum detectable amount (MDA), and some standardization of plating techniques may be needed, but it does not appear that the effort required for these refinements would be excessive.

b. Is Additional Urine Sampling Worthwhile (i.e., Workers at Clean-Up Time)?

The environmental monitoring program should be accelerated if there is a lot of dusty (i.e., earth moving) work planned and this should identify any significant expected increase in airborne contamination. If the environmental monitoring confirms a sharp increase in airborne activities, a resumption of the urine sampling and in-vivo measurement program should be considered. However, a significant upward trend should be confirmed first since the inhalation dose has not been a major contribution to the total dose commitment.

It seems unlikely that the ingestion dose would be significantly altered by dusty operations. Probably the environmental monitoring

efforts to track any buildup of cesium concentration in food stuff would serve as the best trigger to resume urine sampling.

4. Suggestions for Obtaining Funding Support

I offer no suggestions for alternate source of funding for either the environmental or bioassay monitoring programs.

If an autopsy study were undertaken, it is possible that the DOE Office of Health and Environmental Research might consider providing some support as the data would provide useful information beyond the scope of the Marshall Island Program.

5. Is it Important to Pursue Autopsy of Marshall Island Population Group?

The data that would be collected from an autopsy study would be useful to evaluate the effectiveness of the bioassay program in estimating committed doses. If the participants were representative of those showing the higher excreta levels of plutonium in urine, it could effectively establish a reasonable upper bound for plutonium burdens (and thus dose) for the occupants. To date, very little human data are available for chronic exposure to plutonium since most plutonium intakes occur as a result of accidental releases. The data collected could also advance our knowledge of any age bias on the distribution of plutonium in the body organs as the result of age at the time of intake. Even a relatively small group could provide useful information since the mode of intake would be the same for all participants, which is not usually the case for intakes by occupational workers in plutonium facilities.

6. Are Doses Due to Alpha as Important as Doses from Gamma Emitters (i.e., What Should be Used the 50-Year Committed Effective Dose Equivalent or the 50-Year Committed Organ Dose Equivalent)?

Both the 50-year committed effective dose equivalent and the 50-year committed organ dose equivalents need to be estimated (or perhaps even 70-year committed doses). Probably the organ doses will prove to be the more useful for this population group to correlate with specific types of cancer, should there be any excess develop.

It is my understanding that organ doses due to alpha radiation are quite important, but I am certain there are many others (such as Bill Bair) who are much more qualified to comment on this than I am.

7. General Comments

The approach taken by Brookhaven to estimate doses appears to be generally well conceived. In reviewing their estimates of annual intakes for several of the Marshallese listed in the tables they made available to us at the meeting it seemed to me that they have been quite conservative; that is they tended to overestimate the intakes and thus the resulting dose. When the BNL estimates were compared to estimates based on the Gen-Mod Computer Program which incorporates ICRP model without any modifications whatsoever, the BNL estimates were on the average 20% higher than the Gen-Mod estimates. Even with the conservative approach taken by BNL only a few of the reported intakes appear to be statistically positive, that is above background. Several of the others appear to be possibly positive and some of these may be confirmed by additional sampling. In reviewing the data provided, it seemed unusual that of the adult males included in the table, those with longer exposure period (>3 years) tend to have received smaller intakes. It is possible that this is an artifact and is due to some condition that is not obvious at this time. Though the BNL estimates may not prove to be too accurate since they are based on intakes, it is my opinion that their study has already demonstrated that the doses from plutonium (both inhalation and ingestion) are insignificant.

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THE UNIVERSITY OF UTAH

November 12, 1986

Mr. Harry U. Brown
Assistant to the Manager For Off Continent Operations
Nevada Operations Office
U.S. Department of Energy
P.O. Box 14100
Las Vegas, Nevada 89114

Dear Mr. Brown:

My review of the Marshall Islands dosimetry programs is enclosed. This letter summarizes my specific comments relevant to the five issues you wished us to address.

The LLNL dose commitment methodology in my opinion is conservative for Cs-137 (i.e., likely overestimates dose) and is realistic for Pu-239.

It is certainly true that the dose from Pu in diet is small relative to that from Cs-137.

I have compared the results of the LLNL prospective estimates as amount of Pu delivered to the bloodstream against the retrospective estimates based on BNL measurement of urine samples, and found the two differed by less than a factor of three for five of the longest post test residents of Bikini. This difference is rather small considering the potential for error in both approaches. The retrospective technique has large uncertainties associated with the application of urinary excretion functions for time post exposure exceeding 1 year and the analytical methodology is still developmental, not yet well documented and unchecked against the technique in other hands; although it may be quite correct.

The BNL technique should be supported to fruition as it has application not only to monitoring accumulation of Pu in urine, but also to research. The technique should receive peer review and be developed to the point of being routinely accepted.

Pu should be considered in the resettlement options, but is clearly less important than Cs-137 given current estimates of Cs-137 dose. If Cs-137 exposure can be reduced an order of magnitude, the Pu would in a relative sense become more important.

However, I believe the resultant options as currently conceived should be

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based primarily on estimation, limitation and management of the dose for Cs-137.

It is very important to continue the development of the Pu in urine techniques and also to find innovative ways to interpret the results in terms of systemic intake of Pu.

I believe additional urine sampling of workers doing cleanup is warranted and desirable. It may be possible to obtain fundamental information on the excretion rate of Pu in man for times greater than 200 days post exposure if such workers are properly monitored and for a sufficient time.

Funding should be made available from the agencies responsible for the weapons testing program, either the Division of Military Applications of the DOE or the Department of Defense. I prefer the former because of their track record in managing good scientific work in the environmental area with applied problems of human exposure.

It is extremely important to obtain a limited number of human bone or liver samples to help decide whether the prospective or retrospective dosimetric models for Pu are accurate. Such samples may be available either from surgery, biopsy, or autopsy. In the continental U.S. femur heads from hip surgery are being used for Pu measurements and a valid data base exists against which to compare such results from the Marshall Islands when and if they become available. The major organs of interest are bone, liver, and lung and even tissue samples with a mass of 0.1 g are readily measureable for Pu using the fission track method. Any autopsy work should be coordinated with The Hanford Environmental Health Foundation's Transuranium Registry. The president of HEHF is Dr. Bryce Brietenstein (509-942-6010).

In addition, in vivo counts for Am-241 should be made on several long term post test occupants of Bikini to aid in assessing the adequacy of the models for the dose and risk assessment.

I would like to submit an analysis of the relative dose estimation methodologies of BNL and LLNL once Pu is introuduced to the bloodstream, but that will not be possible for me to undertake before February.

Please be assured that I found both your contractors to be capable, thoughtful, and current with the latest techniques. The differences between them arise from items of genuine scientific uncertainity and possibly also dosimetric models, and are, to the extent I have indicated in the enclosed report, resolvable.

Sincerely yours,

McDonald E. Wrenn

McDonald E. Wrenn, PhD
Professor

MEW/jmb

Encl.

cc F.W. Bruenger

EVALUATION OF THE MARSHALL ISLANDS DOSIMETRY PROGRAM AND DOSE ESTIMATES

McDonald E. Wrenn, PhD
Professor of Pharmacology
University of Utah
School of Medicine

November 12, 1986

INDEX

THE DOSE FROM Cs-137:

The LLNL dose estimates for Cs-137 are significant, most probably significant overestimates, and are controlling for purposes of assessing timing and conditions for resettlement.

The significant pathway is terrestrial foods and, of these foods for an adult female resident of Eneu Island, about 80% would come from coconut fluids and solids (Kuhn aand Dreyer, 1982). Thus the expected dose commitment depends primarily upon the rate of consumption of coconut fluids and solids, and the time rate of decrease of Cs-137 in this foodstuff over decades.

The assumption that Cs-137 disappears from coconuts with a half time equal to the physical decay half time, is conservative. Wrenn et al. showed in Jamaica, that Cs-137 accumulated in dairy milk from farms with underlying limestone based soils, similar to that found in the Pacific atolls, was proportional to cumulative deposition in soil and was declining with a half time of 8 years (Wrenn, et al., "Radioactivity Studies: Cs-137 and Sr-90 in Milk, Grass and Soil From Jamaica," Volume III, Progress Report, AEC contract 30-1 (3086), Institute of Environmental Medicine, New York University).

If one were to use a more realistic half time for environmental availability, the 50 year integral dose estimates for Enjebi residents (Robison, Table 10, NCRP-5) of 15 rems would be considerably lower. For illustrative purposes assuming 10 years for an effective environmental half time would decrease that dose estimate from 15 rems to approximately 6.2 rems.

Thus there is a great incentive to know the effective rate of disappearance of Cs-137 from foodstuffs and not just to assume radiological decay, as this knowledge should greatly affect the decision regarding time and conditions of resettlement.

In addition Robison (personal communication) has indicated that intense fertilization will reduce the Cs-137 content of coconuts substantially, and that this effect is sufficiently persistent in time that repeated fertilization could result in long term reduction in Cs-137 content of coconut fluids and solids.

Thus strictly by managing the content of Cs-137 in coconuts by fertilization, it is possible that resettlement could be made an option in the near future without exceeding an expected average whole body 50 year integral dose of 5 rem.

Even on Bikini Island, it is possible that whole body doses could be kept

below 500 mrem/year even in the absence of imports, if a vigorous and targeted fertilization program were maintained.

Quantitative assessment of the effective rate of decrease of Cs-137 from local foods (particularly coconuts) and the efficacy and persistence of reduction of Cs-137 in local foods (particularly coconuts) by fertilization, deserve very high priority and the results should be included in the estimates of doses expected after resettlement.

THE DOSE FROM Pu-239:

There are at least six steps required to retrospectively assess the dose (and expected biological effects) to man from intakes of Pu using analysis of Pu in urine.

1. Proper sampling of urine (i.e., collection in an uncontaminated environment, complete collections over four or more days, etc)
2. Documentation of opportunity for exposure; primarily time and location
3. Chemical extraction of Pu from urine and fission track analysis and quantification
4. Choice of mathematical models, relating urine excretion to intake
5. Choice of mathematical models relating intake to dose
6. Biological assessment of expected risks (either intake related or dose related)

Item 1: More complete documentation of urine sampling locations, protocols, and subsequent handling is desirable.

Item 2: Again complete documentation of exposure histories by individual is required to better apply models to estimate exposure from the results of urine sampling.

Item 3: See the report from F.W. Bruenger (Appendix B) re chemistry of Pu extraction from urine.

Item 4: See Appendix A for complete remarks. Great uncertainty is introduced by the choice of an appropriate urinary excretion model for Pu for times in

excess of one year past start of exposure, as there is a fundamental gap in our basic understanding about the rate of excretion of Pu from the human body at times exceeding 1 year post intake.

COMPARISON OF LLNL PROSPECTIVE PREDICTIONS AND BNL'S RETROSPECTIVE MEASUREMENTS AND ESTIMATES OF SYSTEMIC INTAKE OF Pu:

		BNL	
Case#	T(d)	Bq	pCi
6019	1800	1.11	30.2
6210	3700	3.22	87.6
966	1500	1.10	29.9
6001	3300	0.11	3.0
6128	3300	2.44	66.4
MEAN	2720	1.60	43.4

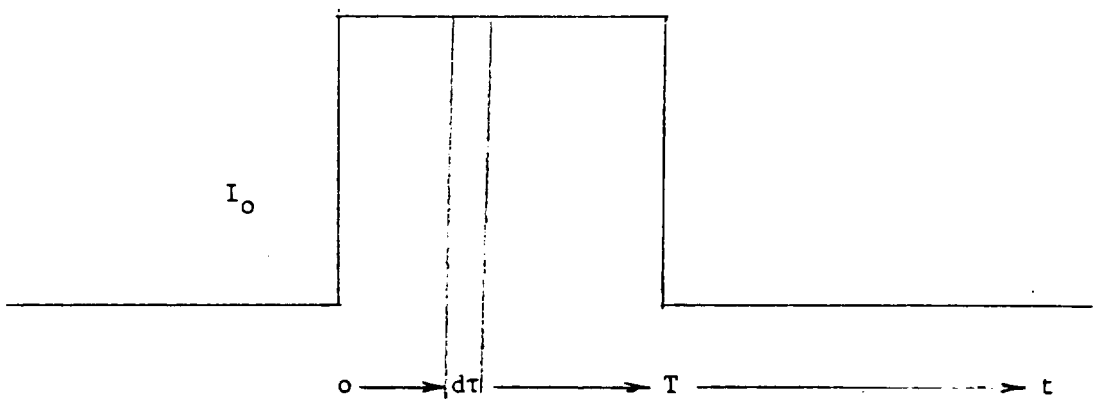
Thus for the 5 longest exposed residents of Bikini Island, the retrospective estimate of intake to blood is 43 pCi, accumulated over 7.45 years on the average, or, neglecting systemic excretion entirely, about 5.8 pCi/y. The results from case 6001 is worrisome because it is an order of magnitude lower than any other result, and the temporal duration of exposure was 9 years, the second longest reported.

To compare with the LLNL model, (see Table 2), I extrapolated the liver and bone burdens to 7.45 years (between their listed 7 and 8 years) and assumed 90% retention in these 2 organs, and that the amount transferred to liver equals that to skeleton (45% each). For a 7.45 year exposure period, the LLNL model of inhalation predicts approximately 18.2 pCi intake to blood. Thus BNL predicts 2.4 times as much intake to blood. The two estimates are not significantly different, given the uncertainties in both. As shown in Appendix A, the choice of which urinary excretion equation to use can affect the estimate of systemic intake by this much. If Lessard et al. used Jone's Equation 10, the prospective and retrospective estimates would be numerically very close. My opinion is that Equation 10 from Jones is a more appropriate mathematical model for the human urinary excretion data and accordingly should be used in the retrospective analysis instead of Equation 9. The inherent uncertainties remain however in the retrospective assessment.

APPENDIX A

ESTIMATION OF PU INTAKE BASED ON URINALYSIS

Consider the case where there is a daily input of Pu to bloodstream, that the rate of input is constant at a daily rate of I_o, that the input continues for a time T, and we want to calculate the fraction of the intake excreted daily in urine at time $t > T$.



$$E = \text{daily excretion} = \int_o^T I(\tau)d\tau \; f(t-\tau)$$

$$= \frac{I_o T}{T}\left(\sum_{i=1}^{4} \frac{k_i e^{-\lambda_i t}}{\lambda_i}\left(1 - e^{-\lambda_i T}\right)\right)$$

$$\frac{E}{I_o T} = \frac{1}{T}\sum_{i=1}^{4} \frac{k_i\, e^{-\lambda_i t}}{\lambda_i}\left(1 - e^{-\lambda_i T}\right)$$

Where k_i and λ_i are taken from the Jones excretion model

Lessard, lists his estimates of total intake of Pu-239 by ingestion and inhalation of 13 people based upon the results of the urine sampling and measurements, and application of a urinary excretion function (Jones et al, 1985) in a table entitled "Results For Adult Males at Bikini Atoll." This table is basically untested in man for the time interval relevant for the urine samples taken here. Lessard's estimates can be used to estimate total intake to blood. The estimate of mean intake to blood is 130 pCi, with a range from 2 to 870 pCi.

I have performed independent calculations (see Table I) and obtain results consistent with Dr. Lessard's (-4% as the average) for intake to blood. Our assumptions may or may not be identical. If I use Equation 10 of Jones, I obtain a higher fractional excretion by 322% than from the use of equation 9; this means the use of Equation 10 would lead to a 3 fold lower estimate of body burden.

Thus the choice of a model of excretion significantly affects the estimate of intake of Pu.

We know accurate excretion rates for adult humans up to 200 days after intake of Pu, but for longer times our basic knowledge of human excretion rates becomes less and less certain with time. We only know rates well in 2 people at about 10,000 days post injection with Pu.

Thus a major need is to more accurately understand fractional excretion of Pu by man.

RESEARCH NEEDED TO EXTEND THE EXCRETION MODEL TO TIMES LONGER THAN 200 DAYS:

The following controlled set of measurements would allow the DOE to (1) obtain such information, and (2) more reliably assess true rates of intake of Pu on Bikini Island.

Send a set of previously unexposed adults to Bikini for up to 3 months (but 100 days maximum). Measure urinary Pu, by fission track assay before, during, and for at least 100 days after exposure. Measure fecal excretion also. Continue followup of the exposed group for at least 3 to 5 years in order to obtain valid periodic excretion data referenceable to the same methods used to obtain human excretion data in the first 200 days post exposure.

VALIDATION OF THE ESTIMATES OF INTAKE FROM URINARY DATA:

1. If subject 2114's intake of Pu-239 was really 870 pCi to blood, then the intake of Am-241 should be from 50 to 80% as high if the intake were from inhalation of soil (see Table 3, Robison, NCRP-5). In that case, the Am-241 would be readily detectable using the thin dual NaI-CoI detectors developed by Laurer at New York University (Wrenn et al, IAEA, Assessment of Radioactive Contamination in Man, pg 595 ff, In Vivo Measurement of Am-241 in Man, 1970.) Dr. Norman Cohen (914-351-4368) at the New York University Institute of Environmental Medicine could arrange the counting, if the subject could be made available. This is an impressive way in which the results of the urinalyis for Pu could be validated (or not).

2. If the retrospective estimate (from urine analysis) of Pu accumulation in Bikini occupants were related approximatley linearly to the time spent on Bikini (post tests), this would lead credence to the belief that the urinalysis results were valid estimates of intake of Pu. Accordingly, I tested Lessard's estimates of intake against length of occupancy (see following pages). There was no significant correlation and the slope of the non-significant correlation was negative; that is, estimates of intake of Pu decreased with increasing residence time (see Appendix C). This in itself does not mean that the urinary results are wrong, as location on the atoll may well be more important than time spent on it. It does suggest that a careful evaluation of the residential history on the atoll of the subjects measured by Lessard is warranted.